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Nuclear Energy Research Initiative
Deep-Burn Molten-Salt Reactors

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Field of Work Element: Engineering or fundamental Science field, F-1. Advanced nuclear energy systems, including components, structure and reactor power conversion cycles/concepts.

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1. Abstract

The molten salt reactor (MSR)—a fluid fuel reactor, operated successfully at ORNL 30 years ago, demonstrated high burn-up and on-line fueling and found a solution to the corrosion issue, but then the MSR research was discontinued. The thinking at the time was that there were too many reactors types being developed, that the resources and talent were spread too thin and that the efforts should be concentrated on one advanced reactor type, the liquid metal cooled fast breeder reactor (LMFBR).

Now, priorities have shifted. Breeding of extra fissile fuel, the primary motivation for the LMFBR development, is no longer as important a goal as it was in the sixties and seventies. Rather, transmutation of the nuclear waste is becoming a major goal. Many fission reactor concepts are being re-examined, with emphasis on passive safety, non-proliferation, waste minimization, resource utilization, and economics. In this light, the demonstrable advantages of a fluid fuel (with extremely high burnup capability, no burnup reactivity swing and on-line removal of poisons and mobile fission products), coupled with recent technical advances in nuclear technology, modeling, and materials science suggest that the timing is right for a fresh look at the MSR. We propose a multi-laboratory/University study of the MSR, with goals to:

- minimize weapons useable material in storage,
- minimize need for high level waste repository space,
- increase the proliferation resistance of nuclear energy
- make beneficial use of spent fuel from LWRs,
- increase resource utilization,
- greatly expand non-carbon based energy (electricity and hydrogen production) at a cost competitive with alternatives.

2. Project Objectives

The Molten Salt Reactor (MSR) was developed in the 1960s at ORNL as a breeder reactor operating on a thorium-²³³U fuel cycle. The fuel is dissolved in a molten salt [see Section 3.1: “Description of the Molten-Salt Reactor (MSR).”]. Since uranium resources were considered very limited, the design goal was to maximize the breeding ratio of the reactor. At that time, non-proliferation was not an issue and the reactor used high-enriched uranium. Safety was important but not considered a major issue. Community-wide, waste management was not considered a significant issue at the time; thus, no significant research was conducted on waste management.

National goals have changed. Safety, non-proliferation, waste management, and economics are major drivers in the design of a modern reactor. The proposed activity will determine whether an MSR can meet these goals and satisfy today’s design criteria. We believe that the MSR could be a superior reactor in each area.

Molten salt reactors (MSR) have the potential of making nuclear energy significantly more competitive than alternative energy sources by virtue of the following characteristics:

1. Relatively low specific capital investment, low fuel cycle cost, no carbon emissions, and high efficiency for converting thermal to electrical energy. These factors should facilitate economic competitiveness with alternative energy options and allow large-scale deployment.

2. Very high fuel utilization and small amount of waste. This facilitates better use of limited natural resources and the burnup of weapons useable materials. It minimizes the needs for repository space by a factor of ten to one hundred, making for a more sustainable energy system and a cleaner world. Molten salt fuel is one of the most suitable forms for refabrication as remote handled-fuel.
3. Superb safety. The option of continually removing fission products and keeping the fissile inventory at a minimum results in enhanced safety from accidents. The MSR is designed to be passively safe independent of size, not just in small sizes. In contrast, for solid fuel reactors to be passively safe they have to be of a relatively small size (typically up to several hundreds of MWe per unit).
4. Supply of high temperature heat (700 °C with conventional design, and higher with new materials) improves overall efficiency and may be suitable for efficient generation of hydrogen. These advantages could stimulate a market place transition away from carbon based fuels.

The MSR concept deserves to be re-evaluated, because it can satisfy today's priorities to:

- minimize weapons useable material in storage,
- minimize need for high level waste repository space,
- increase the proliferation resistance of nuclear energy
- make beneficial use of spent fuel from LWRs,
- increase resource utilization,
- greatly expand non-carbon based energy (electricity and hydrogen production) at a cost competitive with alternatives.

In the proposed NERI project, the objectives are:

- Address and help overcome the potential technical and scientific obstacles to the long-term future use of nuclear energy in the U.S., including the issues involving resistance to proliferation, economics and nuclear waste disposition. The proposed research will make a major contribution to this objective by quantifying the benefits associated with Molten Salt Reactors in each of these areas.
- Advance the state of nuclear technology to maintain a competitive position in overseas markets and a future domestic market. The proposed MSR design will provide a commercially viable, safe, proliferation-resistant path for the US nuclear power industry.
- Promote and maintain a nuclear science and engineering infrastructure to meet future technical challenges. The proposed research will build infrastructure in several laboratories and will also train students in this art.

The proposed research fits well within the scope of the NERI solicitation in two categories:

- **Advanced Nuclear Energy Systems:** This program element includes the investigation and preliminary development of advanced reactor and power conversion system concepts that offer the prospect of improved performance and operation, design simplification, enhanced safety and reduced overall cost. Proposed projects may involve innovative reactor, system or component designs, alternative power conversion cycles for terrestrial applications, or other important design features and characteristics.

- **Advanced Nuclear Fuels/Fuel Cycles:** Research and development is needed to provide measurable improvements in the understanding and performance of nuclear fuel and fuel cycles with respect to safety, waste production, proliferation resistance, and economics to enhance the long-term viability of nuclear energy systems

3. Background

3.1 Description of the molten-salt reactor (MSR)-ORNL Design

In the late 1960s, a conceptual design of a 1000-MW(e) Molten-Salt Breeder Reactor (MSBR) was developed [Robertson *et al.* 1971]. The design characteristics are shown in Table 1, and a schematic of the reactor is shown in Fig 1. The proposed MSR concept is similar to that reactor except for changes in reactor design and the associated fuel-salt processing system to change the proliferation-resistance, safety, and waste- generation characteristics of the reactor. The general plant layout, heat-transfer systems, and power-generation systems for the proposed MSR are similar. However, we are looking at a different salt composition to see how much neutron balance degradation occurs by replacing Li by Na to minimize tritium production and replacing Be by Zr to minimize chemical toxicity. Fig. 1 shows a loop design. Pool designs are possible. The loop versus pool debate has yet to take place for the MSR. The major changes in design are:

- *Uranium-233.* The MSBR fuel used weapons-usable ^{233}U . The MSR fuel will be denatured ^{233}U ; a mixture of thorium and ^{233}U , ^{238}U , and other uranium isotopes.
- *LWR actinide wastes burning.* The MSR core design and salt-processing system will be modified to burn other (LWR) fission reactor actinide wastes
- *Plutonium.* The MSR core design and salt-processing systems will be modified to suppress plutonium production and inventories.
- *Safety.* The MSR design will be modified to address modern safety philosophy and criteria. In particular, it will be designed to be passively safe.
- *Waste management* The MSR design will be modified to minimize the inventory and toxicity of actinides that need be disposed of in the repository.
- *Proliferation resistance.* The MSR fuel cycle will be designed to be as proliferation resistant as practical.
- *Alternative molten salt* We will study the feasibility of using NaF-ZrF₄ as the molten salt; it is free of Be that is chemically toxic material and of Li that generates tritium.

The fuel is a liquid mixture of lithium-7 fluoride, beryllium fluoride, thorium fluoride, and uranium fluorides. During operation, various fission products and actinides also form fluorides in the liquid. Nuclear criticality occurs in the reactor vessel, which contains unclad graphite. The liquid-fuel salt flows upward through vertical channels in the graphite. The graphite slows down fast-fission neutrons and creates a thermal neutron flux. The heat is primarily generated in the liquid fuel. The molten fuel has a high boiling point; thus, the reactor operates at atmospheric pressure. The liquid-fuel salt enters the reactor vessel at 565 °C (1050 °F) and exits at 705 °C (1300 °F). The reactor and primary system are constructed of modified Hastelloy for corrosion resistance to the molten salt. An inert cover gas is used to prevent unwanted chemical reactions. For a discussion of the Hastelloy and the strategy to deal with corrosion see p 81-87 of [Engel *et al.*, 1980]. The strategy was to add 1 to 2% Nb to the Hastelloy and to keep the salt

reducing by maintaining the ratio of UF_4/UF_3 less than 60. This strategy showed good corrosion suppression.

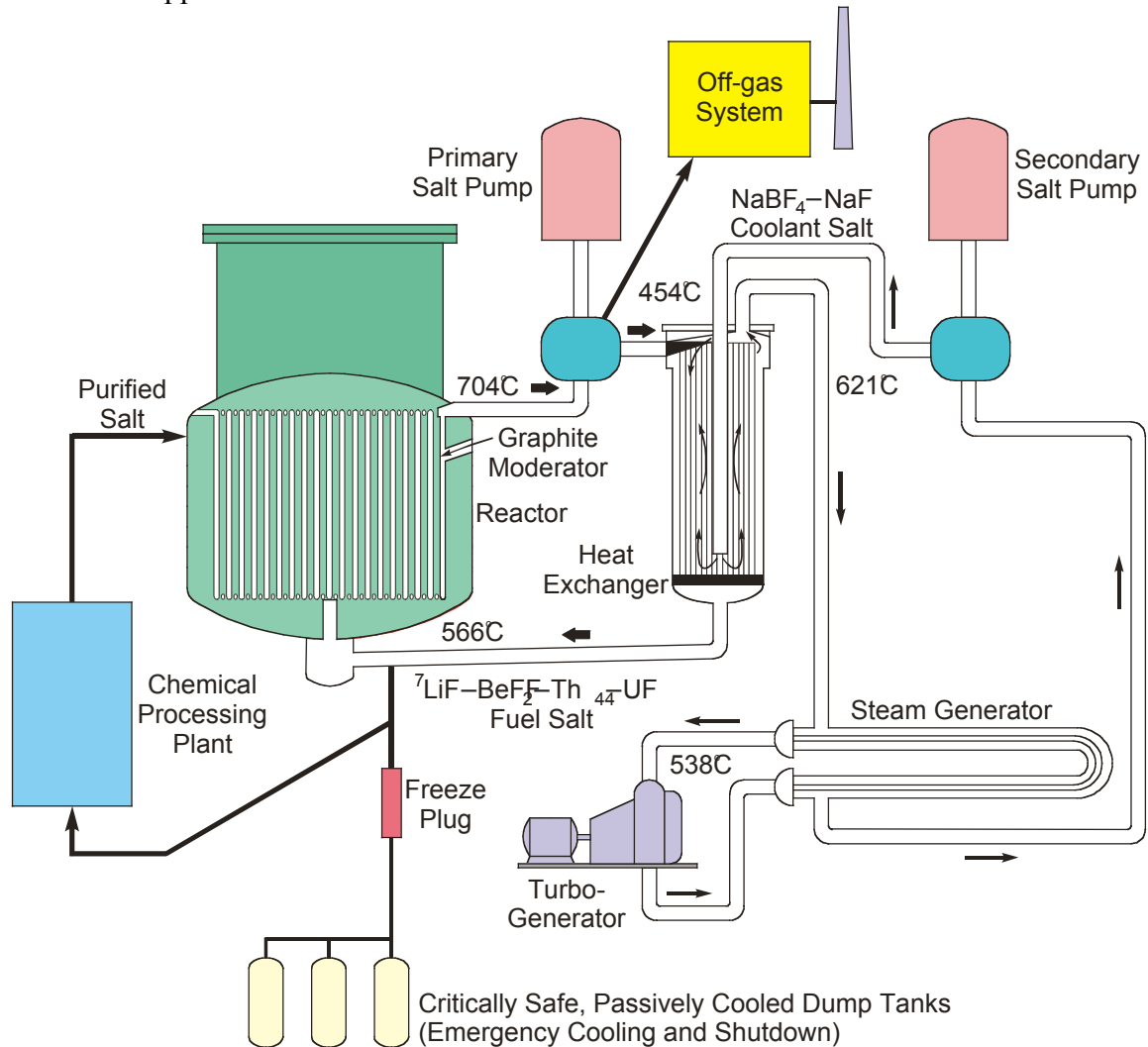


Fig. 1: Schematic of the ORNL molten-salt reactor. Adding ${}^{238}\text{U}$ to the thorium can make this reactor proliferation resistant.

The fuel flows to a primary heat exchanger, where the heat is transferred to a secondary fluid. The liquid fuel flows back to the reactor core. The secondary fluid (NaBF_4-NaF) provides isolation between the molten fuel and the steam cycle and a means to trap the small amounts of tritium (~ 2400 Ci/d) that may be generated in the primary coolant. The heat-transfer fluid flows to a steam generator to produce steam and back to the primary heat exchanger. A conventional steam cycle converts the heat to electricity. The electrical efficiency of the plant is $\sim 44\%$. The high efficiency, as compared to that of LWRs, is a consequence of the high reactor operating temperatures and is a nice advantage of the MSR. The temperatures are determined by the need to ensure low salt viscosity and a significant margin between the salt melting point and the system operating temperature. It is a consequence of the selection of the salt composition. Xenon and other fission-product gases are stripped from the salt in the primary-system

circulation pumps. The reactor has control rods for rapid shutdown, however, during normal operation, the control rods are in the fully withdrawn position. . A very strong negative temperature coefficient of reactivity was demonstrated during operation of the Aircraft Reactor Experiment and the Molten Salt Reactor Experiment [Haubenreich, 1970], and this feature is a fundamental characteristic of a molten-salt-fueled reactor. As salt is heated, it expands and fuel is removed from the reactor.

The 700°C molten salt outlet temperature could be raised somewhat, possibly permitting gas turbine power conversion systems and hydrogen production, however neither is the objective of this proposal.

Table 1: Characteristics of a Large MSR

| | | | |
|-------------------------|---|-----------------------------------|--|
| Net electric generation | 1000 MW | Maximum core flow velocity | 2.6 m/s |
| Thermal efficiency | 44.4 % | Total fuel salt | 48.7 m ³ |
| Core height | 3.96 m | ²³³ U | 1,500 kg |
| Vessel design pressure | 5.2 10 ⁵ N/m ² (75 psi) | Thorium | 68,100 kg |
| Graphite mass | 304,000 kg | Salt components | ⁷ LiF-BeF ₂ -ThF ₄ -UF ₄ |
| Average power density | 22.2 kW/L | Salt composition (see line above) | 71.7-16-12-0.3 mol % |

3.2 Non-proliferation MSR designs

Several limited studies [Bauman, 1977; Engel, 1978] were undertaken to identify methods to improve the proliferation resistance of the MSBR. One study [Engel *et al.*, 1978 and 1980] examined the possibility of an MSBR that operates with isotopically diluted ²³³U. The study indicated that isotopic dilution of ²³³U (<12 wt % ²³³U in ²³⁸U) to a non-weapons-usable material is feasible, but *it did not examine how to reduce the resultant plutonium inventory or the implications of the plutonium isotopics*. Feasibility was defined in terms of maintaining a CR greater than one.

The reactor starts up on low-enriched uranium (LEU). After startup, thorium and depleted uranium are added as needed. The thorium is the fertile fuel to make ²³³U. The ²³⁸U in the depleted uranium is used to maintain the ²³³U as non-weapons usable ²³³U. With a CR of slightly better than 1, there is no need for adding fissile fuel after startup. A small side stream of molten salt is processed to remove fission products (This will minimize parasitic capture of neutrons by fission products, minimize the source term in the core and avoid exceeding solubility limits for fission products in the salt). If the fission product solubility is exceeded, the products can precipitate out and block flow channels. The fuel is never removed from the plant during its lifetime. Lower-cost, inefficient fission-product separations are used because this molten salt (after processing) is immediately mixed back with the molten salt in the reactor. The amount of actinides, carried over with the fission products is desired to be very small, however. Unlike solid fuels, there are no cost or technical reasons for efficient separations because the un-separated fission products are sent back to the reactor.

Actinides never leave the reactor, except in trace amount in the waste streams of FPs, some with the graphite waste and some may be deposited on metallic components, such as steam generators. Other than these small amounts, each actinide is either

fissioned or absorbs a neutron to become a higher actinide. Ultimately, all the actinides that do not get into the waste streams are fissioned. A direct consequence of this fuel cycle is the destruction of nearly all actinides with minimal actinides to the wastes. In terms of waste management, the reactor is a partitioning-transmutation machine, which destroys long-lived actinides; this has major implications for the repository in that repository criticality and safeguards are virtually eliminated by greatly reducing actinides in the wastes. Each MSR will require only 1-10% of the repository needs of today's LWRs of a comparable capacity or other reactors without reprocessing and recycle. This waste reduction is a major thrust of the proposed work.

The fuel cycle has major impacts on the isotopes of the fissile materials in the MSR. The deep burn results in unusual plutonium isotopes. Plutonium-242 becomes the dominant plutonium isotope and ^{239}Pu becomes a minor plutonium isotope in a MSR.

Table 2 shows the expected equilibrium plutonium isotopes for weapons-grade plutonium and plutonium from various reactors. The column *PR-MSR* refers to the calculated equilibrium plutonium isotopes, as determined by [Engel, 1978], for an MSR, which contains ^{233}U and sufficient ^{238}U such as to convert the ^{233}U to non-weapons-usable ^{233}U . That study had as a goal to modify the MSR to make the ^{233}U non-weapons usable. The reactor core design was not modified to minimize plutonium inventories. The last column, *LWR Actinide Recycle in MSR*, is the equilibrium plutonium isotopes for an MSR designed as a partitioning-transmutation machine to burn plutonium and higher actinides from LWR. Greenspan *et al.* (2001) did these calculations. For the proliferation resistant MSR we propose to study, the plutonium isotopes are expected to be between those in the last 2 columns of the table. Plutonium-239, the plutonium isotope preferred for weapons, is a relatively minor plutonium isotope in these MSRs.

Table 2: Plutonium Isotopes, percent

| Isotope | Weapons-grade | Reactor-grade (PWR) | PR-MSR [Engel 1978] | LWR actinide recycle in MSR |
|-------------------|---------------|---------------------|---------------------|-----------------------------|
| ^{238}Pu | | 1.3 | | 2.3 |
| ^{239}Pu | 93. | 56.6 | 30 | 4.5 |
| ^{240}Pu | 6.5 | 23.2 | 18 | 17.9 |
| ^{241}Pu | 0.5 | 13.9 | 14 | 5.0 |
| ^{242}Pu | 0.0 | 4.7 | 38 | 70.2 |

3.3 Molten-Salt Reactor Studies in Japan

The Molten-Salt Reactor Experiment (MSRE) operated successfully for four years in the late 1960s at 8 MWth [MacPherson, 1985]. Should the MSR development get going again it might go through a series of steps such as those advanced by a series of studies in Japan on a small reactor (7 MWe) called mini-Fuji, and a mid-size reactor (155 MWe) called Fuji-II [Furukawa *et al.*, 1992]. These studies are illustrated in Fig. 2 and 3. The Japanese designs are strongly based on the ORNL design. We would base our studies on these designs as well.

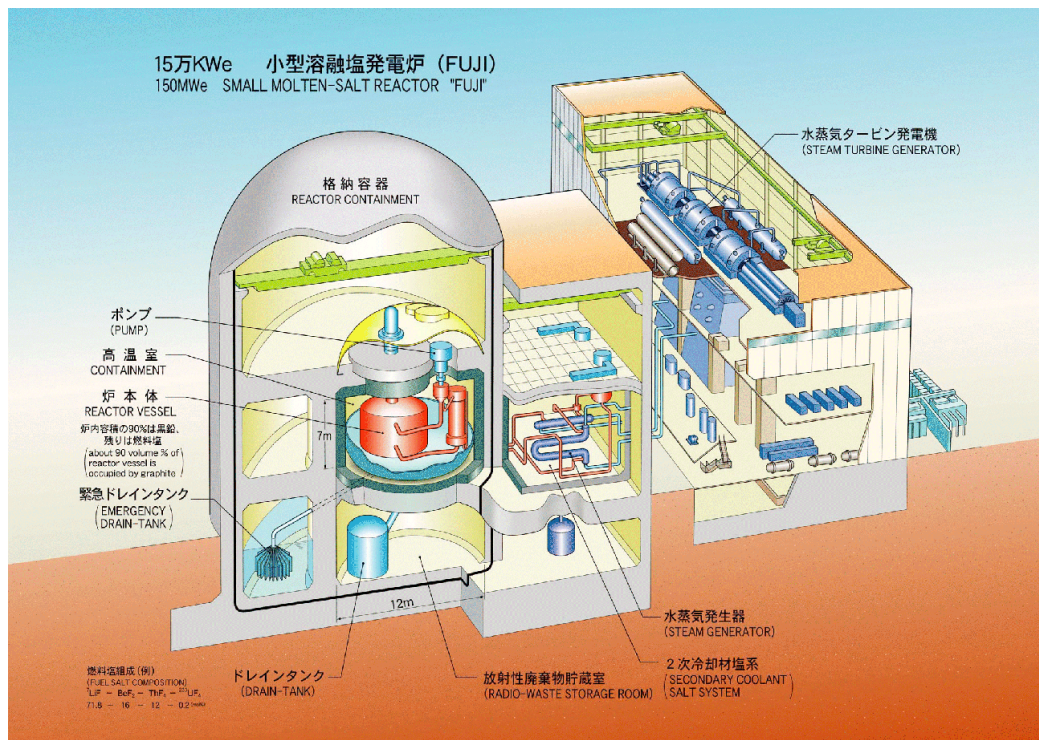


Fig. 2: Molten salt reactor, FUJI (Courtesy of K. Furukawa).

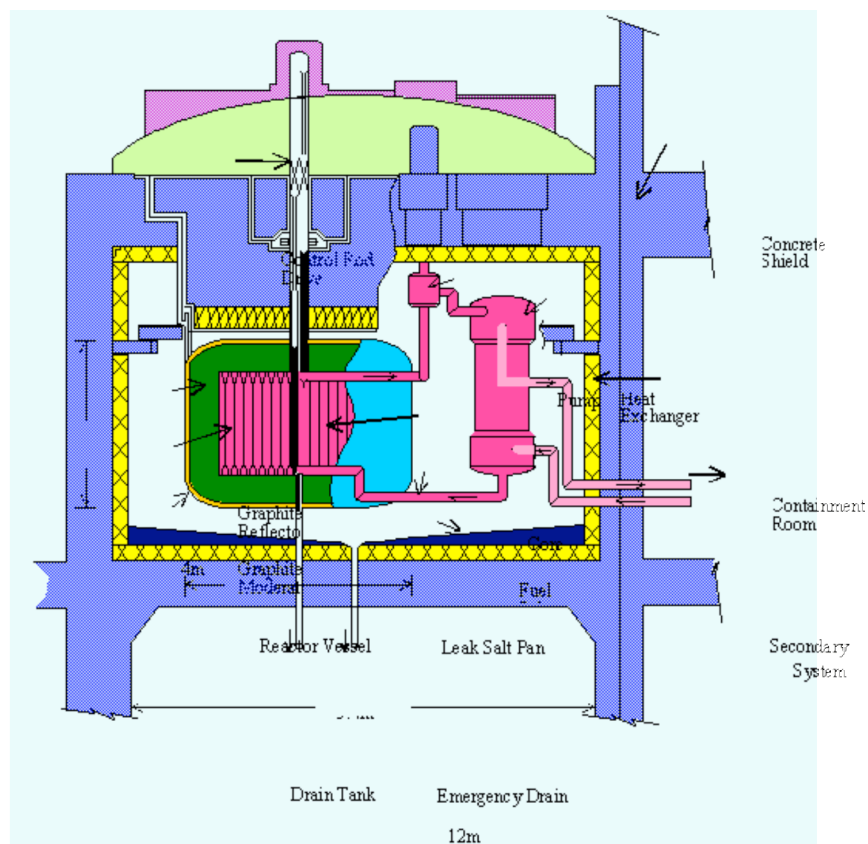


Fig. 3: Molten salt reactor primary system –FUJI (Courtesy of K. Furukawa).

3.4 Cost of Electricity Studies

The cost of electricity from an MSR was recently estimated [Moir, 2002], based on pre-1980 designs. When compared to coal and LWRs of the same era the COE was lower. The ratio of the COE for the MSR to that of the LWR is 0.93. We will see whether this cost advantage still holds when the design is analyzed against revised modern standards. Cost comparisons such as those of [Delene, 1994] should be made.

4. Importance of Proposed Project

Molten salt reactors have the potential of meeting the goals of Generation IV reactors better than solid fuel reactors. They also have the potential of meeting the goals of the high-level waste transmutation program better than solid fuel reactors. In fact, they may enable doing most if not all of the transmutation planned for accelerator-driven subcritical reactors. The proposed study will give DOE the basis for further planning in this area of nuclear research.

Energy R&D planners need to know if nuclear energy can be expanded greatly, be safe, be economical, have adequate resources, have acceptable impacts on waste repositories and have acceptable risks of weapons proliferation. The molten-salt reactor holds the promise of better options for the future. This study will thoroughly assess this option for energy planners to act upon.

Good repository science will result in new robust waste forms and a strategy to minimize repository needs by a factor of ten to one hundred due to application of efficient processing coming out of modern physical chemistry science.

5. Itemized Work Plan

5.1 LLNL

We will evaluate the impact of the MSR on economics, proliferation and national security issues. For example, one basis is to assume 1000 MSR plants are operating by the end of the century. What impact on repository needs for processed LWR spent fuel would be obtained? Much of our findings will be on a per reactor basis. Studies will be carried out to include the impact of changes in design guidelines since the last major work published in [Engel *et al.*, 1980] to include the impact of burning actinides, processing and waste forms. The cost of electricity for MSR of pre-1980 vintage was estimated to be about 90% that of the same era LWR. We will update this estimate by updating all aspects that went into these estimates. The impacts of MSR on repositories (follow-on to Yucca Mountain) under various scenarios will be evaluated.

We will revise the MSR design to meet our project objectives by incorporating new materials, new or appropriate chemical process technologies and items required to meet modern safety guidelines.

Our goals for this study are:

1. Prove that it is possible to design a large MSR to be passively safe
2. Prove that we can design a MSR to be proliferation resistant
3. Prove that we can design MSR to burn most of the actinides from LWR spent fuel and to need a relatively small repository volume
4. Prove that the MSR has the promise to be economically competitive

5. Prove that the overall environmental impact of the MSR is acceptable (or better than that of LWRs)
6. Provide possible scenarios for deployment of MSRs

The LLNL contributions will be made by Ralph Moir, Jim Hassberger, William Halsey and others.

5.2 INEEL -- Safety and Environmental Issues

The design of the molten salt reactor will be evaluated in view of the changes in safety standards and methodology since the molten salt reactor days of two or more decades ago and in view of the increased actinide inventories in some of the burning scenarios.

The Molten Salt Reactor Experiment was shut down about 30 years ago. The shutdown was not because of safety concerns or significant technical issues, but was to focus limited available talent and funding on liquid metal reactors. Since then the emphasis has shifted from high breeding ratio for a plutonium economy to passive safety, non-proliferation, resource utilization, waste handling, synthetic fuel manufacturing, and competitive economics. Our understanding of the phenomena has improved greatly, and better codes have been developed to model neutronics, thermal hydraulics, probabilistic risk analysis, and environmental impact. After TMI and Chernobyl the operating procedures, safety regulations, licensing requirements, and environmental requirements have also changed. In view of the changed emphasis, capabilities, and rules, we need to re-examine the molten salt reactor concept, to see what role it could play in the new world scenario. The preliminary answer to these questions appears to be favorable.

We will do the following tasks:

1. Consider the safety and environmental **philosophy** prevalent in the USA, and how they have changed since the 1960s, and their impact on future power plant designs and licensing compared to the pre-1980 MSR design.
2. Study activation of structure and control of **dose rates** to plant personnel.
3. Estimate the **routine emissions** to environment of noble gases, tritium, and other volatile materials, and discuss how to keep them within safe limits.
4. Estimate the volume of **routine periodic waste** disposal during normal operation.
5. Study a variety of possible **accident scenarios**, including
 - Initiating events
 - Failure of vessel or piping
 - Passive safety features
 - Engineered safety features (if needed)
 - Effects of air or water ingress
 - Source term issues
 - Potential for escape of radionuclides
 - Potential offsite consequences of various scenarios
 - Potential to avoid the need for an offsite evacuation plan
 - Cleanup procedures after an accident
 - Time delay to restart.
6. Evaluate the potential utilization of some separated **fission products**, and the storage requirements for others.

7. Assess the vulnerability of fissile isotopes to **diversion**, including scenarios for theft and countermeasures.
8. Assess the vulnerability of radionuclides in core and in storage to **dispersal** by terrorists, including scenarios and countermeasures
9. Assess the life-cycle **waste products** and discuss their disposal.
10. **Compare** MSR safety and environmental features with other reactor types
11. Identify key safety and environmental **issues** requiring new research and development.

We know *qualitatively* that there are many benefits of MSRs relative to other fission power plants:

- reliable low pressure operation
- no solid fuel fabrication
- online refueling
- negative temperature coefficient
- negative void coefficient
- low radioactive source term
- potential for large unit size
- thorium resource utilization
- high fuel burnup
- high temperature and thermal efficiency
- LWR actinide burnup
- proliferation resistance
- low HLW mass and repository requirements
- low capital cost.

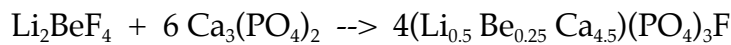
The proposed research will *quantify* those benefits and compare them with other reactor types. Past MSR research did not examine in detail the potential benefits that are relevant to today's priorities, especially safety, nonproliferation and waste transmutation. Thus, the proposed research will be almost entirely new. We anticipate that the results of our studies will make a compelling case for further development of MSRs. The INEEL contributions will be made by T. J. Dolan and others.

5.3 ANL

The MSR waste streams and waste form processing requirements for the fluoride salt systems described above will be evaluated and the requisite processing steps and equipment will be defined. This will include the evaluation of the engineering feasibility of processing methods for the extraction of fission products from molten fluoride salts followed by their subsequent immobilization into waste forms suitable for repository disposal. Substituted fluorapatite (SFA) is a potential waste form host that has been demonstrated on the laboratory scale for LiF-BeF₂ waste salts as part of the MSRE remediation project [Lexa, 1999]. SFA is a naturally occurring mineral that is found to contain ancient actinides in mineral deposits. Fluorapatite is a calcium-based mineral [Ca₅(PO₄)₃F] and Substituted Fluorapatite loaded with the LiF-BeF₂ waste salt was made with the salt ions substituting for calcium [e.g., (Li_{0.5} Be_{0.25} Ca_{4.5})(PO₄)₃F].

The loaded SFA material is made at a relatively low temperature (~500°C). For LiF-BeF₂, the approach involved treating the salt to bring it as close to the composition Li₂BeF₄ as possible and the mixing it with tricalcium phosphate [Ca₃(PO₄)₂] according to

the reaction:



and the product is a ceramic powder. Further lab-scale tests will be carried out to extend this approach to the proposed NaF-ZrF₄ salt system.

Process flow-sheets will be developed and proof-of-principle experiments will be performed. The inventory of actinides and fission products ending up in the waste streams will be quantified and characterized. The strawman path forward for the immobilization of the loaded SFA powder is to follow the method successfully developed for sodalite waste forms for chloride salt. That is, the SFA powders would be bonded with ~25 volume percent glass using a pressureless consolidation method to form a ceramic monolith. The impact of this approach on processing, waste volume, and fuel cycle costs will be estimated.

Itemized tasks for this portion of the project are given below:

1. MSR Process Salt Waste Stream Definition

- *Develop waste stream composition for process flowsheets in consultation with other team members.*
- *Evaluate possible waste fission product separation and immobilization schemes for inclusion in the waste process flowsheet (with ORNL).*

2. Waste Form Processing development

- *Lab-scale experiments to validate SFA as a host for NaF-ZrF₄ waste salts.*
- *Waste form fabrication development*

3. Waste Processing Equipment Definition and Cost Estimates

- *Processing method and equipment selection*
- *Process throughput, scale, and capital cost estimates*

The ANL contributions will be made by S. M. McDevitt and others.

5.4 ORNL

Feasibility of on-line removal of the fission products will be analyzed and recommendations for the proper staging of developments will be made. The batchwise treatment of spent fuel salt to support salt recycle and minimize waste volumes will also be explored. ORNL will lead this effort with assistance from ANL in the areas of electrometallurgical treatments and waste form development. We will examine and assess the processes for removal of fission products with careful attention to low fractional carry over of actinides in the waste stream. The cost of processing will be roughly estimated and the needed development program steps outlined. We will look into the feasibility of using solvents that avoid Li and Beryllium. The MSR produces ~2400 Ci/d of tritium, mostly from neutron reactions on lithium. It is also desirable to explore options that eliminate the chemical toxicity of beryllium (if the nuclear performance is not degraded too much). Alternate fluoride salt systems which meet these objectives will be recommended on the basis of a physico-chemical screening for acceptable high-temperature properties. Special attention will be given to solubility characteristics that can impose burnup limitations (usually trivalent constituents). Minimizing the fission product inventory in the core will improve the neutron economy, will enable reduction of the actinide concentration in the MS, will increase the attainable discharge burnup and will improve the reactor safety. The latter is a direct consequence of a reduction in the source term, in case of an accident.

Previous MSR fluoride separations chemistry was largely directed to support breeding in Th-based systems. Most efforts supported maximizing the neutron economy for this purpose. Spent fuel treatments to support a sustainable disposition of fission products and actinides were not developed. This state of affairs is undeniable, and the experience with the spent fuel from the Molten Salt Reactor Experiment is evidence of the relatively immature state of science in this area relative to the demands of a modern fuel cycle.

Because of the very limited solubility (and corrosion) of fluoride salts in aqueous systems, non-aqueous treatments are required. Some areas of non-aqueous treatments are well developed (fluoride volatility), while others still require considerable research and development (electrochemical, high-temperature treatments). No single separations technology can accomplish the goals necessary to achieve the desired outcome. A careful integration of non-aqueous separations tools will be required. Some new avenues will need to be explored. The need for actinide/lanthanide separation is such a strategic area, and it has recently been suggested that the thermodynamics for electrochemical separation of lanthanides from actinides in fluoride media is more favorable than in chlorides [Prusakov, 1999]. The development of the fluoride analog to the Russian chloride electrochemical fuel cycle for vipac-oxide fuels [Bychkov, 1999] will also be evaluated for its potential to optimize fuel treatments.

Itemized tasks for this portion of the project are given below:

1. On-line Salt Treatment, Fuel Treatment Basis, Alternate Salts
 - *Analysis of on-line salt treatment – needs & capabilities, including cost.*
 - *Analysis of spent fuel treatment flowsheet options based upon modern technology (with ANL).*
 - *Physicochemical screening (solubility, melting point, vapor pressure, etc.) of alternate fuel salt systems that avoid Li (tritium production) and Be (chem. Toxicity).*
2. Non-Traditional Treatments, Definition/Analysis of Reference Flowsheet
 - *Analysis of non-traditional spent-fuel treatment operations (with ANL).*
 - *Definition and detailed analysis of reference spent fuel treatment flowsheet.*
3. Technology Development Plan
 - *Definition of technology development plan to support commercial spent fuel treatments (with ANL).*
 - *Definition of technology development plan to support on-line fuel salt treatments.*

The ORNL contributions will be made by D. F. Williams and others.

5.5 UC. Berkeley

Tasks by UC Berkeley can be divided into two items:

- Optimization of MSR core design (by Greenspan and others), and
- Repository-capacity analysis (by Ahn and others).

5.5.1 Optimization of MSR Core Design

Optimizing the heterogeneous core design. The optimization goals will be to maximize the discharge burnup and minimize the volume and toxicity of the high level waste while being able to maintain criticality and to be below the solubility limit of actinides and fission products in the MS. At least two different MS materials will be

considered: NaF-ZrF₄ and ⁷LiF-BeF₂. The fuel feed besides LWR actinides will be enriched uranium and thorium. The graphite structure will have to be replaced from time to time due to radiation damage. The graphite lifetime in the MSR will be determined as part of the core design.

In order to find the best way to burn LWR wastes, we will look at cases with considerable LWR waste feed and cases with only a little LWR waste feed along with thorium and ²³⁵U feed.

The primary thrust of this task is to find practical and safe core designs that will meet the project goals. There will be six parts to this task: (1) Development and benchmarking of computational tools. (2) Establishment of database and of design constraints. (3) Neutronic parametric studies of cores for the MSR that is to be fuelled with the trans-uranium isotopes from LWR spent fuel and, possibly, Th, using two different solvent molten salts: ⁷LiF-BeF₂ and NaF-ZrF₄. (4) Neutronic parametric studies of cores for the MSR that is to be fuelled with denatured ²³³U and Th. (5) Reference core designs, taking into account thermal-hydraulics and safety considerations. (6) Study of the approach to equilibrium.

5.5.1.1 Computational Tools

Two computer code systems will be used for this study: the SCALE-4.4 code package [SCALE, 1995] and MOCUP [Moore, *et al.* 1995]. We have used both code systems for previous studies of MS reactor cores that are to establish an equilibrium fuel composition [Hughes, *et al.*, 1993][Lowenthal, *et al.*, 2001]. For the first study [Hughes, *et al.*, 1993] we worked out a special sequence of selected modules and data libraries of the SCALE-4.1 computer code package to simulate a MSR core that has a continuous feed in of fuel and continuous extraction of stable and short lived fission products [Shayer, *et al.*, 1994]. For the present study we will implement a similar sequence within SCALE-4.4 – the most updated version of the SCALE code package. The data libraries of SCALE-4.4 use more accurate evaluation of cross-sections (based on ENDF-B/VI) and have a finer energy group structure. Also to be modified will be the definition of the elements to be fed-into, and to be extracted from the MS.

The MOCUP code system [Moore, *et al.* 1995] is presently in use at UCB for the neutronic analysis of thermal as well as fast reactor cores having fixed fuel. It has been thoroughly benchmarked [Briesmeister, 1997] and found to be reliable. It is a linkage code that couples MCNP [Briesmeister, 1997] -- a generalized-geometry, point-energy Monte Carlo transport code, and ORIGEN-2 [Croff, 1980] -- an isotope generation and depletion code. This combination of codes is very useful for depletion and transmutation analysis of systems that have complex geometry or of systems for which ORIGEN-2 does not have sufficiently accurate effective one-group transmutation cross sections. Such cross sections can be generated by MCNP starting from the most updated evaluation of point-energy cross-sections. Before applying MOCUP to the analysis of the MSR we'll have to set ORIGEN-2 to handle continuous feed and continuous extraction of selected elements. Also, special algorithms will have to be added to account for loss of a fraction of the delayed neutrons due to fuel recycling outside of the core and for the extraction of ²³³Pa and feeding back its ²³³U decay product. Doppler broadened cross sections for certain actinides will have to be added to the currently available MCNP library.

The two code systems will be benchmarked against each other for infinite unit cells

of MSR having different MS channel diameter, different lattice pitch, different fuels and different feed and extraction scenarios. The SCALE-based MS reactor analysis code will be the primary tool for scoping studies; solving the neutron transport equation deterministically it is significantly faster than MOCUP that solves the neutron transport equation stochastically. MOCUP, on the other hand, can be used to simulate axially finite unit cell compositions, 3-D cores, control rods and other 2-D and 3-D geometries.

5.5.1.2 Establishment of Data Base and of Design Constraints

The database to be compiled includes the temperature dependence physical properties of the molten salts to be considered, of graphite and of the metallic structural material to be used. Particular attention will be devoted to the search for information on the solubility limits of different actinides and fission products. Also to be determined is the fraction of fission products that can be extracted from the MS and the fraction of actinides that will get into the waste stream in the MS recycling system.

Constraints the study needs to abide by include solubility limits of actinides and fission products, radiation damage limits of graphite, maximum acceptable MS pumping power and maximum acceptable MS flow velocity.

5.5.1.3 Parametric Study of MSR Fuelled with Transuranics from LWRs

The parametric study will be similar to that we recently performed [Lowenthal, *et al.*, 2001] for studying the maximum possible fractional transmutation of different actinides in MS reactor that operates with a once-through fuel cycle. The MS considered in that study was NaF-ZrF₄. The fuel feed consisted of Pu and Minor Actinides (MA) from LWR spent fuel. The variable parameters of the study are the pitch of the MS channels, the diameter of the fuel channels, the volumetric feed and removal rate of the fuel salt, and the concentration of actinides in the feed salt. The core is modeled as an infinite array of heterogeneous, hexagonal cells, (see Fig. 4) unit-cells that are finite in the axial direction; thus accounting for the effect of axial reflectors, shields and axial neutron leakage.

The search for the equilibrium concentration is done iteratively; for a given concentration of actinides and fission products in the MS, MCNP [Briesmeister, 1997] eigenvalue calculation is performed. In addition to the eigenvalue k (multiplication factor) we extract from the MCNP run the total neutron flux in the MS, effective one-group spectrum averaged cross sections for all of the core constituents, and the flux of neutrons having energy above 10 keV in the graphite sleeve. Using these one-group cross-section and total flux, a set of coupled steady-state rate equations (Eq. 1) are solved to obtain a new equilibrium concentration of the actinides. If the actinides concentration significantly differs from that used for the MCNP calculation, a new iteration of MCNP run followed by solution of the rate equations is performed until convergence is reached.

$$\begin{aligned} 0 &= \sum_i \sigma_{i \rightarrow A} \phi N_i / 2 + \sum_j \lambda_{j \rightarrow A} N_j - \lambda_A N_A - \sigma_A \phi N_A / 2 + F_A - R N_A \\ 0 &= \sum_i \sigma_{i \rightarrow B} \phi N_i / 2 + \sum_j \lambda_{j \rightarrow B} N_j - \lambda_B N_B - \sigma_B \phi N_B / 2 + F_B - R N_B \end{aligned} \quad (1)$$

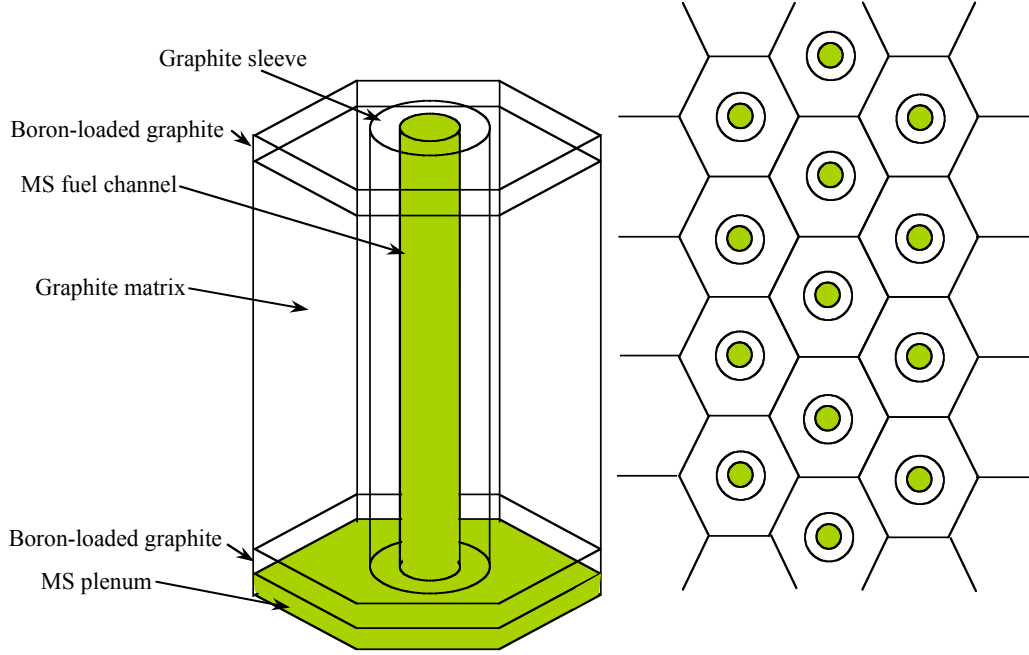


Fig. 4: Configuration used to model with MOCUP the graphite-moderated molten salt reactor

In Eq. 1, N_A is the molar concentration of isotope A in the reactor's stock of molten salt, λ_A is the decay constant for A , $\sigma_{A \rightarrow B}$ is the spectrum-averaged (one-group) microscopic cross section for the reaction transmuting a nucleus of A into a nucleus of isotope B , ϕ is the total average neutron flux in the fuel, F_A is the rate of feed of A per unit volume of molten salt in the reactor system (perfect mixing is assumed), and R is the fractional rate of removal of molten salt from the reactor system. The reaction rates in Eq. 1 are divided by 2 to account for the fraction of time the salt is outside of the core while delivering heat to the heat exchangers

Figure 5 illustrates selected results obtained [Lowenthal, et al., 2001] from such a parametric study for a NaF-ZrF₄ MS. The solubility limit of actinides in this MS is estimated to be 1.56 mole %, at a MS temperature of 550°C. It is found that it is possible to reach an equilibrium core composition in which the actinides are below their solubility limit and k_{eff} is close to 1.0. With the particular design parameters considered for the study summarized in Fig. 5, k_{eff} is ~ 0.97 ; assuming that the radial non-leakage probability is ~ 0.97 . Nevertheless, by adjusting the power density, MS flow rate and concentration of actinides in the MS feed it appears possible to achieve an equilibrium state that is critical.

5.5.1.4 Parametric Study of Denatured ²³³U-Th Fueled MSR

This study will be similar to that described in Sec. 3 but will consider denatured ²³³U-Th fuel cycle. The denatured ²³³U will contain primarily ²³⁸U ($\geq 80\%$). It is envisioned that this denatured uranium will be obtained from reactors that burn Pu and MA in the presence of Th and a suitable amount of ²³⁸U.

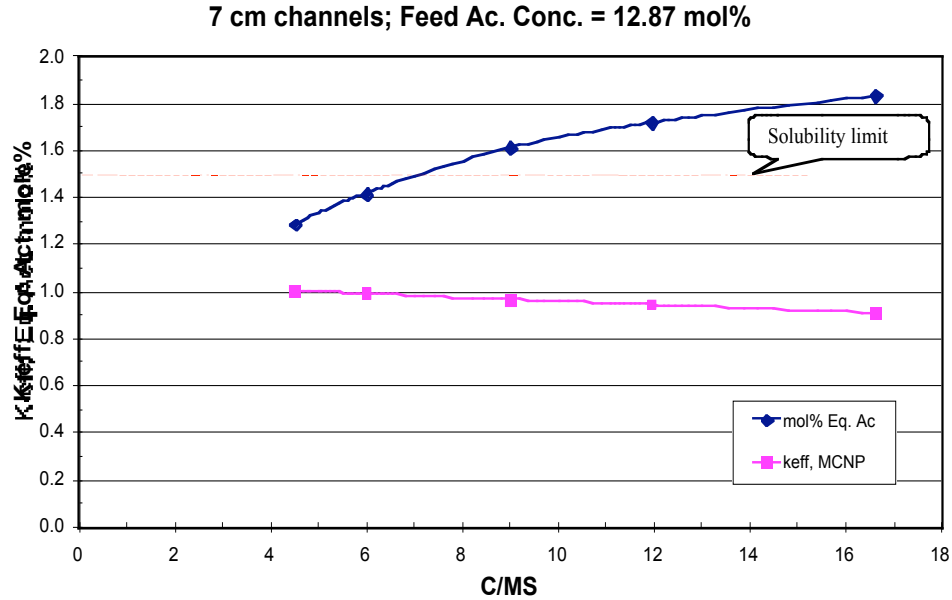


Fig. 5: Dependency of k_{eff} and Actinides (Ac) equilibrium concentration on graphite-to molten salt volume ratio for 7-cm fuel channel diameter at a constant MS feed of 0.8 liters/day having Actinides concentration of 12.87 mol%

5.5.1.5 Reference Core Designs

Focusing on the promising design domains of critical equilibrium MS cores as identified using the simplified model described in Secs. 2 and 3, we'll carry-out a detailed three-dimensional neutronic analysis for a small number of MSR cores. This detailed analysis will be done using MOCUP. It will account for all the actinides and fission products. The ORIGEN2 part of MOCUP will be set to simulate continuous feed and continuous removal of MS.

Among the parameters to be calculated are the power density distribution, flux levels and radiation damage rates, temperature coefficients of reactivity and void coefficient of reactivity. The control system of the reactors will be designed to provide adequate shutdown margin. Also to be estimated is the MS pumping power requirements and the inventory of actinides and fission products that will end up in the high-level-waste stream.

Accident scenarios will be identified and preliminary analyzed. A passively safe design will be attempted that is based on the thermal expansion of the MS. The thermal expansion will dispel actinides with the MS. A design will be searched in which this MS expansion will have a significantly negative reactivity effect.

5.5.1.6 Approach to Equilibrium

The preceding tasks consider MSR in which the fuel has reached its equilibrium composition. In the present task we'll study the evolution of the fuel composition towards its equilibrium state. Among the questions to be addressed is what is the optimal strategy for fueling the reactor and how long will it take to approach the equilibrium composition. The time evolution of the fuel isotopics will be determined.

One of the fuel cycle options to be considered during the approach to equilibrium is the addition of thorium to the Pu and MA from LWR spent fuel as the fuel feed for the MS reactor. This will enable to use the excess neutrons in the pre-equilibrium stage to convert thorium into ^{233}U . The ^{233}U , that will be denatured with ^{238}U , could be used to fuel MSRs that operate on the denatured ^{233}U -Th fuel cycle.

5.5.2 Repository-Capacity Analysis

Theoretical studies will be performed to confirm that repository capacity required for the disposal of the waste from MSR is smaller by a factor of 10 to 100, compared with the conventional LWR spent fuel disposal. This should be performed, based on the waste compositions and solidification (to be studied by ANL and ORNL). Significant reduction in repository space requirement is essential for sustainable deployment of the MSR system at a large scale. It is imperative to demonstrate feasibility of reduction by a factor of 10 to 100 and its implication to the sustainability of the system.

Repository performance should be evaluated from many viewpoints, such as radiological health impact to the public, criticality safety, proliferation resistance, institutional control, cost, public perception, and so on. With the proposed MSR system, it is expected that the amount of actinides to be included in the waste stream would be one to two orders of magnitude smaller than that in the conventional LWR spent fuel. In the past repository performance studies, however, it has been claimed that the mass of actinides in the waste does *not* affect the repository performance because of the low solubility of actinides in groundwater. Recently, it was pointed out by the team of UC Berkeley researchers [Ahn, *et al.*, 2002] that even under the solubility-limited condition, spatial configuration of the canister array in a repository could have an important influence on the concentration of radionuclides leaving the repository area, thus affecting the amount of radionuclides to be released to the far field around the repository.

In the proposed project, utilizing the waste compositions and solidification determined by ANL and ORNL, performance assessment and preliminary design study will be made for a geologic repository. A hypothetical repository is considered. The computer code, VR, for repository performance assessment is readily available at UC Berkeley [Tsujimoto, *et al.*, 2000]. The code utilizes the Parallel Virtual Machine (PVM) technology to perform radionuclide transport calculations for multi-canister configurations.

For this task, three steps will be taken:

(1) While ANL and ORNL are working to develop a reference-case specification for the waste from MSR, UC Berkeley will work on necessary extension of the VR code to accommodate specific features of the MSR waste. This can be completed in the first fiscal year of the project. Also in the first year, measures for the repository performance will be developed. Presently, the radiological exposure dose rate to the public is considered to be the most important measure, which is primarily determined by the concentration of radionuclides in groundwater. Because the efforts to be made in this project is to reduce the mass of long-lived actinides in the waste, developing alternative and/or additional measures for the repository performance that show sensitively the effects of mass reduction would be essential.

(2) In the second fiscal year, a thorough performance assessment study will be made with the MSR waste specifications developed by ANL and ORNL. Effect of waste

arrangement in a hypothetical repository is studied. Through this study, feedback would be made to the designs of the reactor and the partitioning processes to enhance the sustainability of the system (i.e., to decrease the repository space requirement).

(3) In the third year, iteration between the front-end and the back-end of the system will be performed, to tune the system design. We will revisit the performance measures and check if these are properly set.

6. Collaboration

Efforts will be made to collaborate with other groups interested in MSR. The Japanese especially have an interest, which will be pursued. Especially K. Furukawa's whose work is illustrated by his references and Fig. 2 and 3. He has agreed to collaborate so his resume is included. The Central Research Institute for Electric Power Industry (CRIEPI) of Japan has been studying molten-salt dry process for recovery of actinides in collaboration with UC Berkeley would participate in the proposed project. Formal agreement is yet to be arranged. French and Russian related work and contacts may develop.

7. Project Schedule and Milestones

The tasks are given in Table 3 in detail for each organization for each of three years. Each organization will generate contribution to a yearly progress report on each of the tasks. Significant overlap will require close coordination. The proposed funding level is given in Table 4.

Table 3: Summary of Tasks

| | First Year | Second Year | Third Year |
|-------|---|---|--|
| LLNL | <ol style="list-style-type: none"> 1. Develop ref. scenarios (size, fuel cycle, objectives) 2. Assess pre-1980 MSR designs 3. Assess impacts of design modernizations 4. Est. performance (cost, repository needs, proliferation) | <ol style="list-style-type: none"> 1. Revise project objectives 2. Revise MSR design for latest materials and chemical processes 3. Est. performance (cost, repository needs, proliferation) | <ol style="list-style-type: none"> 1. Revise project objectives 2. Revise MSR design for latest materials and chemical 3. Est. performance (cost, repository needs, proliferation) |
| INEEL | <ol style="list-style-type: none"> 1. Assessment of design changes to accommodate newer safety rules 2. Safety philosophy 3. Dose rate control 4. Routine emissions 5. Routine waste 6. Accident scenarios | <ol style="list-style-type: none"> 1. Accident Scenarios (continued) 2. Fission product disposition | <ol style="list-style-type: none"> 1. Fissile diversion 2. Terrorists 3. Life cycle wastes 4. Comparison of reactor types 5. Key safety & environmental issues for R&D. |
| ANL | <ol style="list-style-type: none"> 1. Waste Stream Flowsheet Development: Define Waste Composition 2. Review Possible Treatment Methods for Fission Product Extraction including cost estimates. 3. Begin Process Development for substituted fluorapatite (SFA) waste form. | <ol style="list-style-type: none"> 1. Support fission product removal feasibility study through review and experiment. 2. Demonstrate SFA waste form fabrication 3. Characterize SFA waste form. | <ol style="list-style-type: none"> 1. Support fission product removal feasibility study through review and experiment. 2. Evaluate SFA waste form behavior in repository relevant conditions. |

| | | | |
|------|---|--|--|
| ORNL | 1. Analysis of on-line salt treatment – needs & capabilities, including cost estimates. 2. Analysis of spent fuel treatment flowsheet options based upon modern technology. 3. Physicochemical screening (solubility, melting point, vapor pressure, etc.) of alternate fuel salt systems to avoid Li (tritium production) and Be (chem. toxicity). | 1. Analysis of non-traditional spent-fuel treatment operations. 2. Definition and detailed analysis of reference spent fuel treatment flowsheet. | 1. Definition of technology development plan to support commercial spent fuel treatments. 2. Definition of technology development plan to support on-line fuel salt treatments. |
| UCB | (MSR Core Design) 1. Development and benchmarking of computational tools. 2. Establishment of data base and of design constraints. 3. Neutronic parametric studies of MSR cores fuelled with TRU from LWR spent fuel | (MSR Core Design) 1. Optimal reference core design for LWR TRU fueled MSR, taking into account thermal-hydraulics and safety considerations 2. Neutronic parametric studies of cores fuelled with denatured ²³³ U and Th 3. Optimal reference core design for denatured ²³³ U-Th fueled MSR, taking into account thermal-hydraulics and safety considerations | (MSR Core Design) 1. Accident analysis of the reference MSR designs. 2. Criticality safety analysis of the reference MSR designs 3. Study of the approach to equilibrium. |
| | (Repository Capacity) 1. Modification of the VR code to accommodate MSR waste. 2. Development of alternative/additional measures for repository performance. | (Repository Capacity) 1. Repository performance assessment for a reference-case MSR waste. 2. Preliminary design for a geologic repository, in which the repository space required is minimized for the reference-MSR waste. 3. Feedback to FP removal, fuel treatment, and reactor design for further improvement of repository performance and space reduction. | (Repository Capacity) 1. Iteration between the designs for the reactor, the fuel treatment, and the waste treatment, to minimize the repository space requirement. 2. Re-consideration of repository performance measures. |

Table 4: Budget Summary(k\$)

| | First Year | Second Year | Third Year |
|-------|------------|-------------|------------|
| LLNL | 300 | 300 | 300 |
| INEEL | 150 | 150 | 150 |
| ANL | 150 | 150 | 150 |
| ORNL | 150 | 150 | 150 |
| UCB | 169 | 169 | 172 |
| Total | 919 | 919 | 922 |

8. Organizations and Qualifications

Lawrence Livermore National Laboratory will be the principal investigating

organization, with responsibilities for planning and coordinating all work performed under this proposal. Special strengths are in areas of non-proliferation and repository considerations.

Idaho National Engineering and Environmental Laboratory will have responsibility for safety.

Argonne National Laboratory will have responsibility for waste form and processing.

Oak Ridge National Laboratory will have responsibility for processing.

University of California, Berkeley will have responsibility for reactor physics and repository considerations.

Each organization has broad strengths that overlap.

9. Key Personnel

Ralph W. Moir

Education

B.S. 1962--Engineering Physics, University of California, Berkeley

Sc.D. 1967--Nuclear Engineering, MIT, Cambridge, Mass.

Professional Associations, Societies and Honors

Registered Professional Nuclear Engineer in the State of California, NU782.

American Physical Society, Fellow 1981, Plasma Physics Div

American Nuclear Society, Fellow 1989, Fusion Energy Div, Chairman 93 to 94.

Publications

- R. W. Moir, et al., "Tandem Mirror Hybrid Reactor Design Annual Report", Lawrence Livermore National Laboratory, Livermore, CA, UCID-18808 (1980).
- R. W. Moir, "The Fusion-Fission Fuel Factory, Chapter 15, p. 411-451, in *Fusion*, Vol. 1 Part B, edited by E. Teller, Academic Press, New York (1981).
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After his BS degree in Engineering Mechanics, Dr. Dolan served two years in the Navy, then earned a PhD in Nuclear Engineering from the University of Illinois (1970). He spent 10 months as a post-doctoral student at the Novosibirsk State University and Institute of Nuclear Physics, USSR. He served as a professor of nuclear engineering at the University of Missouri-Rolla 1971-1989, where he taught about 20 different courses and served as Department Head 1985-1987. He had summer research jobs at the Lawrence Livermore National Laboratory, Oak Ridge National Laboratory, Los Alamos National Laboratory, and Institut National de la Recherche Scientifique – Énergie, Université du Québec. He was a visiting professor at Tsing Hua University, Taiwan (1977-1978). He was a consultant to Phillips Petroleum Company on the Ohmic Heated Toroidal Experiment (1981-1988). In 1987 he joined the Idaho National Engineering Laboratory and worked on physics applications, fusion reactor design studies, space nuclear power, arms control, and university programs.

From 1995-2001 he served as Head of the Physics Section, International Atomic Energy Agency (IAEA), Vienna, Austria, doing administration, international research coordination, and organization of technical meetings on nuclear fusion research, utilization of research reactors and low-energy accelerators, and nuclear instrumentation. He organized a coordinated research project involving 11 countries on "Comparison of Compact Toroid Configurations" (nuclear fusion research). He presented invited lectures in Iran, Italy, and Japan, and attended or organized technical meetings in Brazil, Canada, China, Croatia, Egypt, Finland, France, Germany, India, Italy, Japan, Korea, Portugal, Russia, Slovenia, and the Ukraine. In September 2001 he returned to the Idaho National Engineering and Environmental Laboratory, where he has been involved with Generation-4 reactor studies, advanced fuel studies, fission product chemistry, national security programs, and university programs.

Selected Publications

- T.J. Dolan, *Fusion Research*, Pergamon Press, 1982 (textbook, now available on CD free from the IAEA: n.pawel@iaea.org)

- J. K. Hartwell, L. Forman, and T. J. Dolan, "Warhead Demilitarization - Some Pros and Cons," *Verification Technologies Review*, 2, No.6, Nov/Dec 1990.
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- T. J. Dolan, "Possible generation of self-magnetic fields," *Fusion Science and Technology* 40, 119-124 (September 2001).

Sean M. McDeavitt

Institution: Argonne National Laboratory

Classification: Nuclear Engineer/Section Manager, Materials Development Section

Specialty: Nuclear Materials and Materials Processing

Education:

| <u>Degree</u> | <u>Discipline</u> | <u>University</u> | <u>Date</u> |
|---------------|---------------------|-------------------|-------------|
| Ph. D. | Nuclear Engineering | Purdue University | 1992 |
| M. S. | Nuclear Engineering | Purdue University | 1990 |
| B. S. | Nuclear Engineering | Purdue University | 1987 |

Experience Summary:

Dr. McDeavitt has been developing processing methods for nuclear materials for over ten years. He joined the Chemical Technology Division (CMT) of Argonne National Laboratory in December of 1992. He developed a stainless steel-15 wt. % zirconium (SS-15Zr) waste form alloy to immobilize radioactive metallic waste from an electrometallurgical processing method used to treat spent fuel. He led the SS-15Zr project from concept to demonstration as a group leader in the Waste Form Development Section of CMT; this alloy waste form is now being demonstrated in the Fuel Conditioning Facility at ANL-West (near Idaho Falls, ID). In addition, he is currently working with an external contractor to develop advanced crucible materials for melting reactive liquid metal alloys. He also serves as a project manager for a 1999 NERI project for the development of an advanced thorium oxide-based cermet fuel and he has studied swelling and densification mechanisms in nuclear fuel. Dr. McDeavitt is a member of the Minerals, Metals, and Materials Society (TMS) (Chairman, Reactive Metals Committee) and the American Nuclear Society (Member, Materials Science and Technology Division Executive Committee).

Selected Publications:

- S. M. McDeavitt, "Uranium Processing for the Nuclear Fuel Cycle," *J. Miner. Met. Mater.*, **52** (2000) 11.
- S. M. McDeavitt, D. P. Abraham, and J. Y. Park, "Evaluation of Stainless Steel Zirconium Alloys as High Level Nuclear Waste Forms," *J. Nucl. Mater.*, **257** (1998) 21.
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Dave F. Williams

OAK RIDGE NATIONAL LABORATORY

Education

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|----------------------------------|------------|----------------------------------|
| Virginia Institute of Technology | B.S. 1978 | Chemical and Nuclear Engineering |
| University of Tennessee | M.S. 1985 | Chemical Engineering |
| University of Washington | Ph.D. 1991 | Chemical Engineering |

David F. Williams has 15 years of professional experience in radiochemical R&D. His experience has ranged from design work in support of production of sol-gel particulate nuclear fuel, to development of flowsheet and equipment for the radiochemical recovery of special isotopes, to more basic chemical research. For the past three years he has led the basic research studies that established the salt chemistry necessary for the remediation of the Molten Salt Reactor Experiment at ORNL. He is the present Group Leader of the Chemistry Research Group in the Chemical Technology Division.

Selected Publications

- D. F. Williams, A. S. Icenhour, L. D. Trowbridge, G. D. Del Cul, and L. M. Toth, "Radiolysis Studies in Support of the Remediation of the Molten Salt Reactor Experiment," Transactions of the American Nuclear Society (invited paper published in 1999 Winter Meeting Proceedings, November 14–18, 1999, Long Beach, California).
- D. F. Williams, G. D. Del Cul, and L. M. Toth, "Molten Salt Fuel Cycle Requirements for ADTT Applications," *3rd International Conference on Accelerator Driven Transmutation Technologies and Applications (ADDTA '99)*, Prague, Czech Republic, June 7–11, 1999, (paper We-I-17) in http://www.fjfi.cvut.cz/con_adtt99/.
- D. F. Williams, J. Brynestad, "Evaluation of Fluorine-Trapping Agents for Use During Storage of the MSRE Fuel Salt," ORNL/TM-13770, Oak Ridge National Laboratory, Oak Ridge, Tennessee, May 1999.

- D. F. Williams, L. M. Toth, and G. D. Del Cul, *Chemical Interactions During Melting of the MSRE Fuel Salt*, ORNL/M-5506, Oak Ridge National Laboratory, Oak Ridge, Tennessee, November 1996.
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- D. F. Williams, G. D. Del Cul, and L. M. Toth, *A Descriptive Model of the MSRE after Shutdown Review of FY95 Progress*, ORNL/TM-13142, Oak Ridge National Laboratory, Oak Ridge, Tennessee, January 1996.

Charles Forsberg

OAK RIDGE NATIONAL LABORATORY

Education

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|--|---------------------------------|
| University of Minnesota, Minneapolis | B.S.—1969, Chemical Engineering |
| Massachusetts Institute of Technology, Cambridge | M.S.—1971, Nuclear Engineering |
| Massachusetts Institute of Technology, Cambridge | Sc.D —1974, Nuclear Engineering |

Professional Activities and Affiliations

Fellow, American Nuclear Society.
 Member, American Association for the Advancement of Science
 Member, American Institute of Chemical Engineers
 Member, Materials Research Society
 Member, U.S. Department of Energy ²³³U Team
 Member, U.S. Department of Energy High-level Waste Technical Advisory Panel
 Registered Professional Engineer (State of Tennessee)
 Principal holder of eight U.S. patents

Highlights

Dr. Charles Forsberg is a senior staff member of ORNL. His research areas are advanced reactors and fuel Cycles. His doctorate thesis was on uranium enrichment technologies, and he has done subsequent research on reprocessing, fuel fabrication, and other fuel-cycle technologies. He has been the program manager for several programs, including the developmental LWR program, which examined inherently and passively safe LWRs. He holds eight patents in the areas of passive safety systems for power reactors, reprocessing, and waste treatment.

At ORNL, he is a member of the DOE ²³³U multi-site team addressing ²³³U safety and storage issues. He directed the technical studies on disposition options for excess ²³³U. He participated in the DOE TOPS workshops to examine how to improve proliferation resistance in the nuclear fuel, is the U.S. molten-salt reactor contact for the DOE/Russian Proliferation-Resistant Nuclear Technology (PRNT) program, and is a member of the Non-classical reactor team for the Generation IV road map activity. Dr. Forsberg led the team that developed the technical basis for defining weapons-usable ²³³U (>12 wt % ²³³U in ²³⁸U), which is based on isotopic composition. He also developed the

methodology to define waste thresholds for ^{233}U , that is, the concentration of ^{233}U in waste at which safeguards may be terminated because the ^{233}U is practicably unrecoverable. He is currently conducting studies on the future uses of ^{233}U for reactors and other applications. Consequently, reviews of worldwide activities in these areas are being completed.

Selected Publications (Total list is greater than 150 articles and reports)

- C. W. Forsberg, "Are Chemically Separable Weapons-Usable Fissile Materials a Characteristic of Nuclear Power Systems", *Science and Global Security*, (Submitted)
- C. W. Forsberg, "What is Non-Weapons-Usable Material?," p. 62 in *Trans. 1999 Winter American Nuclear Society Meeting, Long Beach, California, November 14–18, 1999*, Vol. 81.
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- C. W. Forsberg, E. C. Beahm, L. R. Dole, A. S. Icenhour, S. N. Storch, L. C. Lewis, and E. L. Youngblood, *Disposition Options for Uranium-233*, ORNL/TM-13553, Oak Ridge National Laboratory, June 1, 1999.
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- C. W. Forsberg, "Plutonium Futures," *MIT Nuclear Systems Safety Course*, Department of Nuclear Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts, July 23, 1998.
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- C. W. Forsberg, C. M. Hopper, J. L. Richter, and H.C. Vantine, *Definition of Weapons-Usable Uranium-233*, ORNL/TM-13517, Oak Ridge National Laboratory, Oak Ridge, Tennessee, March 1998.
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- C. W. Forsberg and A. M. Weinberg, "Advanced Reactors, Passive Safety, and the Acceptance of Nuclear Energy," *Annual Rev. of Energy*, **15**, 133–152, 1990.
- C. W. Forsberg, et al., *Proposed and Existing Passive and Inherent Safety-Related Structures, Systems, and Components (Building Blocks) for Advanced Light-Water Reactors*, ORNL-6554, Oak Ridge National Laboratory, Oak Ridge, Tennessee, December 1989.
- C. W. Forsberg, "Passive Emergency Cooling Systems for Boiling Water Reactors (PECOS-BWR)," *Nucl. Technol.* **76**, 185, January 1987.

- C. W. Forsberg, "A Process-Inherent Ultimate Safety Boiling Water Reactor," *Nucl. Technol.* **72**, 121–134, February 1986.

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Education:

- 1957-1961 B.Sc in Mechanical Eng. + Nuclear Option (Cum Laude), Technion - Israel Institute of Technology.
- 1961-1963 M.Sc in Nuclear Science & Eng., Technion, Israel. "Optimization of the Nuclear Design of a 125 MWe Heavy-Water Natural Uranium Power Reactor."
- 1963-1966 Ph.D in Nuclear Science & Eng., Cornell University, Ithaca, N.Y., USA. "Theory and Measurement of Neutron Importance in Nuclear Reactors."

Relevant Experience:

Ehud Greenspan is a full-time faculty member at UC Berkeley since 1992. He teaches reactor theory and reactor design & analysis courses. Prior to joining UC Berkeley he was an Associate Director for Research and Development at the Nuclear Engineering and Applications Division of the Israeli Atomic Energy Commission. He has extensive and broad research experience in advanced nuclear reactors and nuclear fuel-cycle conception and analysis. He was the PI on dozens of advanced nuclear systems conception and analysis. Among these are studies of molten-salt reactors - see publications No. 8, 9, 20, 22 and 25. He has more than 350 publications a sample of which follows:

1. Ehud Greenspan, "Optimization of the Nuclear Design of a 125 MWe Heavy-Water Natural Uranium Power Reactor," M.Sc. Thesis, Israel Institute of Technology, 1963.
2. E. Greenspan, K. B. Cady and J.P. Howe, "Economic Potential of Variable Enrichment Fuel Elements for Power Reactors," Trans. American Nuclear Society, **9**, 295-296 (1966).
3. E. Greenspan, "Energy Dependent Fine Structure Effects on the Reactivity Worth of Resonances," Proc. Advanced Reactors; Physics, Design and Economics, (J. E. Kallfeltz & R.A. Karam, Ed.) Pergamon Press, pp. 196-205, 1975.
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8. J. Hughes, I. Soares, E. Greenspan, W.F. Miller and Z. Shayer, "Molten Salt Critical Reactors for the Transmutation of Transuranics and Fission Products," Proc. of the GLOBAL'93 International Conference, Seattle, WA, Sept. 12-17, 1993. pp. 644-651.
9. Z. Shayer, J. Hughes, I. Soares, E. Greenspan and W. Miller, "Modifying SCALE-4.1 for Transmutation Calculations," Trans. Israeli Nuc. Soc., 18, VII 27-VII 30 (1994).
10. E. Greenspan, "BWR Fuel Assembly Having Oxide and Hydride Fuel," US Patent No. 5,349,618, Sept. 20, 1994.
11. J. Vujic, E. Greenspan, S. Slater, T. Postam, L. Leal, Greg Casher and I. Soares, "Development of Coupled SCALE 4.2/GTRAN2 Computational Capability for Advanced MOX Fueled Assembly Designs," Proc. Int. Conf. on Math. & Computations Reactor Physics and Environmental Analysis. Portland, OR. April 30-May 4, 1995. pp. 1001-1010.
12. N.E. Brown, J. Hassberger, E. Greenspan and E. Elias, "Proliferation Resistant Fission Energy Systems," Proc. Global'97: International Conf. On Future Nuclear Systems, Yokohama, Japan, October 5-10, 1997. pp. 879-884.
13. T.H. Kim, N.Z. Cho. And E. Greenspan, "Fuel-Self-Sufficient Heavy-Water Lattices for Proliferation Resistant Multiple Fuel Recycling," Trans. Am. Nucl. Soc., 77, 108-110 (1997).
14. E. Greenspan, E. Elias, W.E. Kastenberg and N.W. Brown, "Compact Long Fuel-Life Reactors for Developing Countries," Proc. 9th International Conference on Emerging Nuclear Energy Systems, ICENES'98, Herzlia, Israel, June 28 - July 2, 1998. pp. 74-83.
15. E. Greenspan, W.E. Kastenberg, N.Z. Cho, T.H. Kim and S.G. Hong, "Multi-Recycling of Spent Fuel with Low Proliferation Risk" Proc. 9th International Conference on Emerging Nuclear Energy Systems, ICENES'98, Herzlia, Israel, June 28 - July 2, 1998. pp. 455-464.
16. N.Z. Cho, S.G. Hong, T.H. Kim, E. Greenspan and W.E. Kastenberg, "Fuel Self-Sufficient and Low Proliferation Risk Multi-Recycling of Spent Fuel," Proc. 13th KAIF/KNS Annual Conf., Seoul, Korea, April 1998. pp. 417-425.
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20. M.D. Lowenthal And E. Greenspan, "Parametric Studies For Optimization of a Graphite-Moderated Molten Salt Transmuter," Proc. of GLOBAL-2001, Paris, France, September 2001. 8 pages.

21. E. Greenspan, N. W. Brown, M. D. Carelli, L. Conway, M. Dzodzo, Q. Hossain, D. Saphier, J. J. Sienicki, D. C. Wade, "The Encapsulated Nuclear Heat Source Reactor for Proliferation-Resistant Nuclear Energy," Proc. of GLOBAL-2001, Paris, France, September 2001. 8 pages.
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| B.S. | Nuclear Engineering, University of Tokyo, 1981 |
| M.S. | Nuclear Engineering, University of Tokyo, 1983 |
| Ph.D. | Nuclear Engineering, University of California, Berkeley, 1988 |
| D.Eng. | Nuclear Engineering, University of Tokyo, 1989 |

Joonhong Ahn won Junior Scientist Fellowship from the Japan Society for the Promotion of Science (JSPS) (1988-90). Dr. Ahn joined the faculty in the Department of Nuclear Engineering, University of Tokyo as an Assistant Professor (1990-1993). He moved to Department of Nuclear Engineering, Tokai University (1993-1995). In January 1995, he joined the faculty at Berkeley.

His research interests include the performance assessment of deep geological disposal systems for high-level radioactive wastes (HLW), especially analyses of mass transport through engineered barriers and the natural geological barrier. He is also interested in future of nuclear energy in Asia/Pacific region.

Professor Ahn served as a member of the Planning Committee for the Atomic Energy Society of Japan (1992-1995). He served as the Editor for Radioactive Waste Research (1994-1996), a journal of the Division of Radioactive Waste Management, Atomic Energy Society of Japan. The journal was established when he was serving as the Secretary General for the Division (1993-1995). He serves as a member of the

committees of Technical Journals and Book Publishing of the American Nuclear Society (since June, 2001)

Selected Publications:

- Ahn, J., Y. Furuhashi, Y. Li, and A. Suzuki, Analysis of Radionuclide Transport Through Fracture Networks by Percolation Theory, *Journal of Nuclear Science and Technology*, **28**(5), 433—446, 1991.
- Ahn, J. and S. Nakayama, Modeling for Migration of A Redox-Sensitive Radionuclide in Engineered Barriers, *Nuclear Technology*, **97**(3), 323—335, 1992.
- Ahn, J., and A. Suzuki, Diffusion of the $^{241}\text{Am} \rightarrow ^{237}\text{Np}$ Decay Chain Limited by Their Elemental Solubilities in Artificial Barriers of High-Level Radioactive Waste Repositories, *Nuclear Technology*, **101**(1), 79—91, 1993.
- Ahn, J., S. Nagasaki, S. Tanaka, and A. Suzuki, Effects of Smectite Illitization of Transport of Actinides Through Engineered Barriers of HLW Repositories, 18th International Symposium on the Scientific Basis for Nuclear Waste Management, Materials Research Society, Atomic Energy Society of Japan, October 1994, Kyoto, Japan, 1995.
- Ahn, J., Transport of Weapons-Grade Plutonium and Boron Through Fractured Geologic Media, *Nuclear Technology*, **117**(3), 316-328, 1997.
- Ahn, J., Integrated Radionuclide Transport Model for an HLW Repository in Water-Saturated Geologic Formations, *Nuclear Technology*, **121**(1), 24-39, 1998.
- Ahn, J., Criticality Safety Assessment for a Conceptual High-Level-Waste Repository in Water-Saturated Geologic Media, *Nuclear Technology*, **126**, 303-318, 1999.
- Ahn, J., Preliminary Assessment of the Effects of ATW System Application on YM Repository Performance, *Global '99, International Conference on Future Nuclear Systems*, August 29-September 3, 1999, Jackson Hole, Wyoming (1999).
- Ahn, J., E. Greenspan, and P. L. Chambré, A Preliminary Consideration for Underground Autocatalytic Criticality by Vitrified High-Level Waste in Water-Saturated Geologic Repository, *Journal of Nuclear Science and Technology*, **37**(5), 465-476, 2000.
- Ahn, J., and P. L. Chambré, Alternative Measure for Performance of HLW Geologic Repository, *Global 2001, International Conference on "Back-end of the fuel cycle: from research to solutions,"* Paris, France, 9/9-9/13 (2001).

KAZUO FURUKAWA

Thorium Molten-Salt International Forum

Education

Faculty of science, University of Kyoto

B.S.—1951, Chemistry

Institute for Iron, Steel and other Metals, Tohoku University

D. Sc. —1960, Inorganic Liquid Structural Chemistry

Professional Activities [cf. “Five Hundred Leaders of Influence”(1999), American Biographical Institute]

A visiting fellow at the University of London, Birkbeck College in 1960, he then took the post of associate professor at Tohoku University. He worked at Japan Atomic Energy Research Institute from 1962 to 1983, when he became a professor at Tokai University. He founded the Japanese liquid sodium technology for FBR at JAERI

between 1962 and 1970, and examined several fluid-fuel reactor concepts relating with fission, fusion and spallation. In 1980 he invented the Accelerator Molten-Salt Breeder (AMSB), aiming fissile material breeding, and in 1985 he proposed the simplified small Molten-Salt Fission Power Station named FUJI, which is fuel self-sustainable without continuous chemical processing.

Dr. Furukawa was elected as a foreign member of the Ukrainian Academy of Science in 1991. He was invited by the Elec. de France, President Mr. Bergougnoux in 1987, who decided “no Superphenix No.2”. In 1992 he met with Dr. Allan Bromley, President Advisor of Science and Technology, who encouraged him during the development of the THORIMS-NES (Thorium Molten-Salt Nuclear Energy Synergetics) project, and also gained the support of the following President Advisor of S&T, Dr. John H. Gibbons, in 1997. Dr. Furukawa’s work on the Thorium Molten-Salt Reactor was examined by President Clinton in 1997 and was also presented at the International Conference on Molten-Salt Development at RAND in Santa Monica, California, where he submitted a paper entitled “Conclusions and Recommendations” with 24 peoples from Japan, Russia, Belarus, France, Czech, Turkey and the U.S.A. Three Agency Study (OECD/NEA, OECD/IEA, IAEA) is recommending his work for international developmental item.

Selected Publications (Total list is more than 150 articles and reports)

- K. Furukawa, H. Ohno: "Molten LiF-BeF₂ (Flibe) System", DATA-BOOKS FOR MOLTEN MATERIALS I, Japan Nuclear-Energy Information Center, pp.114 (1980)
- K. Furukawa, Y. Kato, T. Ohmichi, H. Ohno: "The Combined System of Accelerator Molten-Salt Breeder(AMSB) and Molten-Salt Converter Reactor(MSCR)" [Japan-US Seminar on "Th Fuel Reactors"(Oct.1982,Nara)], "Thorium Fuel Reactors", (1985) P.271-281, Atom.Ene.Soc.Japan.
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- K. Furukawa: "Symbiotic Molten-Salt Systems coupled with Accelerator Molten-Salt Breeder(AMSB) or Inertial-Confined Fusion Hybrid Molten-Salt Breeder(IHMSB) and their Comparison," [3rd-ICENES, Helsinki, 1983] Atomkernenergie/Kerntechnik, 44 [1] P.42-45 (1984)
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- K. Furukawa, K. Minami, K. Mitachi, & Y. Kato: "Compact Molten-Salt Fission Power Stations (FUJI-series) and their Developmental Program", Proc.Joint Int.Sympo.Molten-Salts, Proc.Vol. 87-7 P.896-905(1987) Electrochem.Soc.
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- K. Furukawa & A. Lecocq : "Preliminary Examination on The Next Generation Nuclear Reactors in Comparison with the Small Thorium Molten-Salt Reactor", sponsored by Electricite de France (EdF), France [Tokai University

(xiii+100pp)](Dec.,1988)

- K. Furukawa, A. Lecocq, Y. Kato & K. Mitachi :”Summary Report : Thorium Molten-Salt Nuclear Energy Synergetics,” J.Nucl. Sci. & Tech.,Vol.27, No.12, P.1157-1178 (1990)
- K. Furukawa, K. Mitachi, Y. Kato & A. Lecocq :”Global Nuclear Energy System -- Thorium Molten-Salt Nuclear Energy Synergetics—“,Proc.Indo-Japan Seminar on "Thorium Utilization" (Bombay, India, Dec. 10-13, 1990) P.21-28(1991)
- K. Furukawa, A. Lecocq, Y. Kato, & K. Mitachi :”Radiowaste Management in Global Application of Thorium Molten-Salt Nuclear Energy Synergetics with Accelerator Breeders”LA-12205-C Conf. SKN Report No.54 ; UC-940[November 1991] P.686-697.
- K. Furukawa, K. Mitachi & Y. Kato :”Small Molten-Salt Reactors with a Rational Thorium Fuel- Cycle”,Nuclear Engineering and Design, 139, P.157-165 (1992)
- Y.Kato, K.Furukawa, K.Mitachi & S.E. Chigrinov:”Fuel Trajectory in Accelerator Molten-Salt Breeding Power Reactor System including Pu Burning”, "Emerging Nuclear Energy Systems ICENES'93", Ed.H.Yasuda, (1994) P.439-443, World Sci.Pub.
- K.Furukawa: [COMPILED BOOKS] “Important Papers concerning on Thorium Molten-Salt Nuclear Energy Synergetics[THORIMS-NES]”, Vol.I (Oct.,1994), pp.272; Vol.II(Oct.,1994), pp.279. Inst.of Research & Development, Tokai University,
- K.Furukawa, K.Kato & S.E.Chigrinov:”Plutonium (TRU) Transmutation and ²³³U Production by Single-Fluid type Accelerator Molten-Salt Breeder (AMSB)”.AIP CONFERENCE PROCEEDINGS 346 (1995)p.745-751, Am.Inst.Physics.
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- K.Mitachi, Y.Yamana, T.Suzuki, & K.Furukawa :”Neutronic Exam. on Plutonium Transmutation by a Small Molten-Salt Fission Power Station” : IAEA-TECDOC-840, P.183-195(1995)
- K.Mitachi, K.Furukawa, T.Suzuki & A.Namekata:”Pu Burning Molten-Salt Power Station (FUJI- Pu3) for Prevailing Th Nucl.Industry”,ICENES’96:Vol.I(1997)P.180-190, Inst.Phys.Power Eng.
- S.Chigrinov, A.Kievitskaia, C.Rutkovskaia, I.Rakhno, K.Furukawa & A.Lecocq :”Accelerator Molten-Salt Breeder as Fissile Producing Component of THORIMS-NES Concept for Energy Production and Transmutation of Plutonium”.ICENES’96: Vol.II(1997) P.564-571, Inst.Phys.Power Eng.
- K.Furukawa, V.A.Simonenko, K.Mitachi, A.Furuhashi, R.Yoshioka, S.E.Chigrinov, A.Lecocq & Y.Kato:”Thorium Cycle Implementation through Pu-Incineration by Thorium Molten-Salt Nucl. Energy Synergetics”. IAEA Advis.Groop Meet.- Th Fuel Cycle Perspec., April, 1997, Vienna.
- K. Furukawa, Y. Kato & K. Mitachi: “Measure On Global Energy/Environmental Disorder in 21st Century by Thorium Molten-Salt Nuclear Energy Synergetic System [THORIMS-NES]”, [Key Note Address] “Different Disordered-System”, P.1-8 (2001) Ch.Ed. S.K.Srivastava, INDIAS Pub.

10. Facilities and Resources

No special facilities and physical resources are required for the proposed work. However, immediate access to the staffs of each of the participating organizations are very valuable strengths of the proposal, as each is a leader in the fields of nuclear engineering and nuclear weapons.

11. Budget

Table 4: Budget (k\$)

| | First Year | Second Year | Third Year |
|-------|------------|-------------|------------|
| LLNL | 300 | 300 | 300 |
| INEEL | 150 | 150 | 150 |
| ANL | 150 | 150 | 150 |
| ORNL | 150 | 150 | 150 |
| UCB | 169 | 169 | 172 |
| Total | 919 | 919 | 922 |

12. References

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